THE SURFACE TENSION OF THE NEAR-EUTECTIC ALLOYS OF LEAD-BISMUTH SYSTEM

B.B. Alchagirov[1], A.G. Mozgovoy[2], Kh.B. Khokonov[1]
[1] Berbekov Kabardino-Balkarian State University, Nalchik, Russia
[2] Joint Institute of High Temperatures of the Russian Academy of Sciences, Moscow, Russia

Investigating the physicochemical properties of the liquid alloys of Pb-Bi binary system is of major significance in connection with their extensive application in various branches of science and novel engineering. For example, the lead-bismuth eutectic (containing 55.5 mass % or 55.3 at. % Bi [1]) is used as a liquid-metal heat carrier in fast-neutron, nuclear power reactors [2]. Its melting point exceeds but slightly the melting point of sodium, being equal to 397.7±1.5K [3].

An analysis of the investigations of the surface tension (ST) of liquid Pb-Bi alloys [4-6] available in the literature shows that there is a certain discrepancy between the results of these measurements, amounting in some cases to as high as about 5%. Moreover, in study [4] the temperature dependencies of ST for two lead-bismuth melts (50.0 and 56.5 mass % Bi) show a maximum in the vicinity of 470 K, while the experimental data [5, 6] is described by linear approximating equations with negative temperature coefficients.

That is why the authors of this work have investigated the ST of the liquid alloys of a binary Pb-Bi system in the ranges of temperature 473-653 K and concentration 33.3-76.3 mass % Bi. The measurements were performed by the method of a large (sessile) drop on an all-soldered glass instrument [7].

The investigated lead-bismuth alloys were prepared by a weighing method under a superhigh vacuum (about 10^{-5} Pa). And this process was carried out directly in the measuring instrument without opening or unsealing it. The purity of the initial lead and bismuth was not lower than 99.99 mass %, while the contents of gaseous and metallic impurities in each of them did not exceed $1{\cdot}10^{-4}$ and $1{\cdot}10^{-3}$ mass %, respectively. Proceeding in this way, we prepared seven Pb-Bi alloys. Their compositions are given in Table 1. The error in determination of the concentration of components of these alloys was estimated at ± 0.1 mass %.

Table 1. Compositions of the investigated lead-bismuth alloys

Alloy serial number		1	2	3	4	5	6	7
Content of bismuth	Mass %	33.3	38.1	44.5	50.8	64.6	70.4	74.0
	at. %	33.1	37.9	44.3	50.6	64.4	70.2	73.8

In order to reach the equilibrium values of the ST, the investigated lead-bismuth melts were allowed to settle for a period of no less than 1 h. This is necessary due to strong concentration nonuniformity of Pb-Bi alloys prepared by the weighing method which require abnormally long periods of time to establish uniform bulk distribution of their components, even with considerable superheating over the liquidus temperature [8].

Figure 1 illustrates the results of the performed measurements of ST of liquid lead-bismuth alloys. The confidence experimental error, including both the systematic and random components, was found to be approximately 1% with probability of 0.95 in the investigated ranges of temperatures and concentrations.

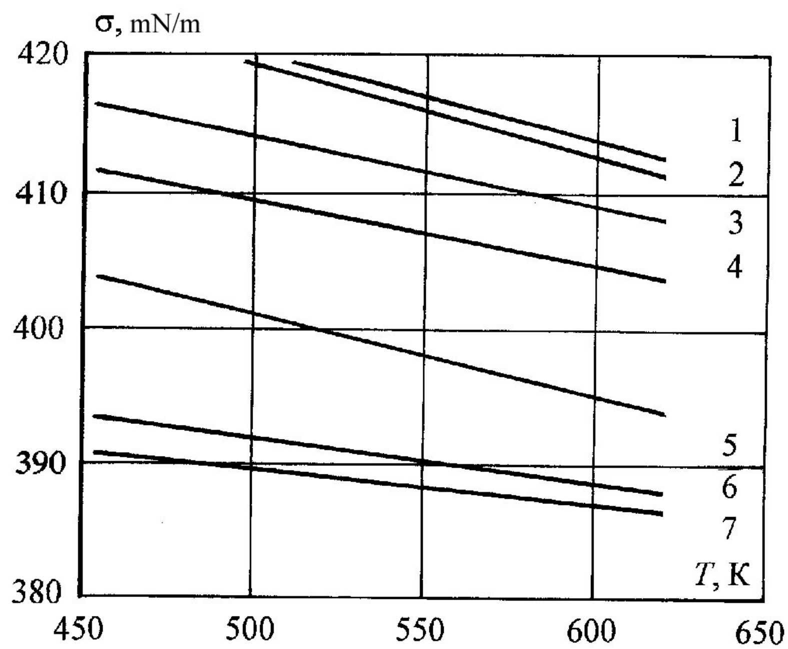

Fig. 1. The temperature dependence of the ST of melted lead-bismuth alloys: 1 – 33.1, 2 – 37.9, 3 – 44.3, 4 – 50.6, 5 – 64.4, 6 – 70.2, 7 – 73.8 at. % Bi.

The obtained experimental data on the ST of molten lead-bismuth alloys was processed by the least-square method using an approximating equation of the following form:

$$\sigma = a_0 + a_1\tau + a_2\tau^2 + \dots . \qquad (1)$$

Here σ is the surface tension, in mN/m; $\tau = T/1000$, and T is the temperature in K, according to the International Temperature Scale of 1990 [9].

The optimum number of the parameters of the approximating equation (1) was found using Fisher' criterion [10]. It turned out that only the coefficients a_0 and a_1 proved to be significant with confidence probability 0.95 for all the investigated Pb-Bi alloys. Their values are listed in Table 2. One can see from this table that in the investigated temperature range the polyterms of the ST of the molten lead-bismuth alloys are linear equations with negative temperature coefficients.

Table 2. The values of the parameters of approximating equation (1)

Alloy serial number	1	2	3	4	5	6	7
a_0	451.9	454.8	438.6	433.1	431.2	408.6	402.1
$-a_1$	63.5	71.8	49.1	47.4	60.2	33.6	24.9

The deviation of the obtained experimental points from the approximating equation (1) with parameters given in Table 2 did not exceed ±0.5%. The latter proves that the systematic error constituted the greater part of the confidence error of the performed measurements.

The experimental data of the present study of the ST of liquid lead-bismuth alloys is in satisfactory agreement with the measurement results of [5] in the comparable range of temperature and concentrations. At the same time, it differs from the experimental data [6] to a considerable extent, and it does not confirm the presence of maxima on the temperature dependencies of the ST of some liquid Pb-Bi alloys, as noted in [4].

In this connection, it should be noted that the discrepancy between the experimental values of various authors on the ST of liquid metals mainly depends on their density values used in calculating ST of metal melts. Unfortunately, such data is unavailable in most works [4, 5] devoted to the investigation of the ST of liquid lead-bismuth alloys. Only in study [6] the authors used their own data on temperature and concentration dependencies of the density of the molten Pb-Bi alloys obtained by the sessile drop method. It is well known [11], however, that this method results in fairly large errors (up to 2%) in the values of the density of liquid metals. This circumstance is probably one of the causes of the discrepancy between the experimental data of this work and those of study [6], which has been mentioned above.

In such cases, the values of the density of the molten lead-bismuth alloys, calculated according to the additivity law, may prove to be more accurate and reliable compared to the results of its experimental investigation, as, for example, in study [6]. This was, in particular, done in [12]. This statement is corroborated by the closeness of thermophysical and physicochemical properties of the components (lead and bismuth) of the Pb-Bi system. The latter also defines small values of the excess volume and the mixing heat of lead-bismuth alloys.

The additivity law for the ST of liquid-metal binary system is written as

$$\sigma = \sigma_1 x_1 + \sigma_2 x_2, \tag{2}$$

where σ_1, σ_2 are the surface tensions of the pure (molten) first and second components of the alloy, respectively; x_1, x_2 stand for the molar concentrations of the first and second components in the alloy, respectively, and $x_1 + x_2 = 1$.

In Fig. 2, the experimental data of this work and study [6] on the ST of liquid Pb-Bi alloys at a temperature of 600K are compared with those calculated according to the additivity law (2). For the latter, the values of the ST of molten lead and bismuth were assumed in accordance with the recommended reference data [12].

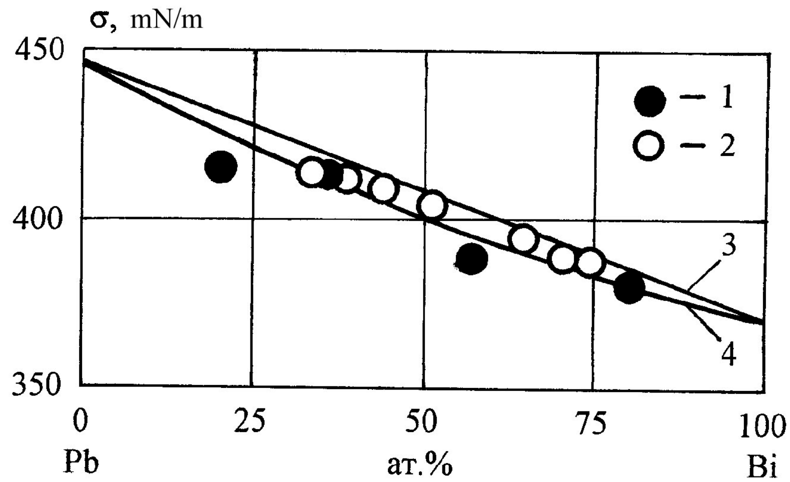

Fig.2. The concentration dependence of the ST of liquid Pb-Bi alloys at 600 K: 1- data [6], 2 – present work, 3 – equation (2), 4 –equation of Nemchenko-Popel [13].

It is apparent from Fig. 2 that the results of this investigation lie systematically lower than the concentration dependence of the ST of lead-bismuth melts defined by the equation (2). These discrepancies, however, are within the limits of the confidence error of the measurements of the ST of liquid Pb-Bi alloys. At the same time, the experimental data [6] deviates from equation (2) to a more considerable extent, and the discrepancy between the two is as high as approximately 15 %. The cause of such discrepancy was discussed earlier.

The results of other measurements of the ST of molten lead-bismuth alloys [4,5] were not included in the above-mentioned analysis. Actually, the experimental data of [4] was obtained in a rather narrow temperature range, while the maximum investigation temperature was 61 K. The experimental data of study [5] is presented in the original publication as graphs plotted on a small-scale drawing, which does not allow to derive the information on the ST of the investigated Pb-Bi alloys necessary for the analysis.

Figure 2 also shows the concentration dependence of the ST of liquid lead-bismuth alloys, calculated using the equation of Nemchenko-Popel [13]. It satisfactorily describes the experimental data of this work, and can be applied to the determination of the ST of liquid-metal binary systems in case the literature lacks corresponding measurement data.

REFERENCES

1. Chirkin V.S. The Thermophysical Properties of Materials for the Nuclear Engineering. Moscow, Atomizdat, 1968. 484 p.
2. Askhadullin R.Sh., Gulevich A.V., Dedul A.V. et al. 50 Jahrs of the Assimilating of Heavy Heat-Transfer Agent Technology. Ed. A.A.Simakov. Obninsk, Kaluga region, Leipunsky Physical and Power Engineering Institute, 2001. 28 p.
3. Kaplun A.B., Shulaev V.M., Linkov S.P., Vartanov Yu.D. The Viscosity of the Eutectic Lead-Bismuth Alloy. <u>The Thermophysical Properties of Substances and Materials.</u>

Novosibirsk, Kutateladze Institute of the Thermophysics of the USSR Academy of Sciences, 1979, p. 105-113.

4. Klyachko Yu.A., Kunin L.L. On the Surface Tension of Eutectic Alloys. Reports of the USSR Academy of Sciences, 1949, v. 64, No. 1, p. 85-86.
5. Pokrovsky N.L., Pugachevich P.P., Golubev N.A. The Investigation of the Surface Tension of Solutions of the Lead-Bismuth System. Russian Journal of Physical Chemistry, 1969, v. 43, No. 7, p. 2158-2159.
6. Kazakova I.V., Lyamkin S.A., Lepinskikh B.M. The Density and the Surface Tension of Melts of Pb-Bi System. Russian Journal of Physical Chemistry, 1984, v. 58, No. 6, p. 1534-1535.
7. Alchagirov A.B., Alchagirov B.B., Khokonov Kh..B. The Device for the Determination of the Surface Tension of Metallic Solutions in Complete Concentration Range of Compositions. Russian Federation Patent No. 2086957, 1999.
8. Kaplun A.B. On the Motives of Physical Properties Anomalies of Metallic Melts. Proceedings of Russian High Educational Institutions. Ferrows Metallurgy, 1985, No. 7, p. 30-35.
9. Preston-Thomas A. The International Temperature Scale of 1990 (ITS-90). Metrologia, 1990, v. 27, No. 1, p. 3-10.
10. Hudson D.J. Statistics. Lectures on Elementary Statistics and Probability. Geneva, Switzerland, 1964. 257 p.
11. Alchagirov B.B., Dadashev R.Kh. The Large Drop Method for the Determination of the Density and the Surface Tension of Metals and Alloys. Nalchik, Berbekov Kabardino-Balkarian State University, 2000. 94 p.
12. Kirillov P.L., Denickina N.B. Thermophysical Properties of Liquid-Metal Heat-Transfer Agents (Reference Tables and Relationships). Review FEI-0291. Moscow, ZNIIAtominform, 2000. 42 p.
13. Nemchenko V.P., Popel S.N. The Dimension Estimation of Adsorbing Particles onto Isotehrms of the Surface Tension and the Density. Russian Journal of Physical Chemistry, 1969, v. 43, No. 7, p. 1822-1828.